

(translation)

## **(12) Japanese Patent Laid-Open(A)**

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(54) Title: Composition for electrolyte, electrolyte, method of producing thereof, and battery

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(57) Abstract:

PROBLEM TO BE SOLVED: To provide a composition for an electrolyte, capable of showing high film-forming properties and high ion conductivity, the electrolyte and its manufacturing method, and a battery using it.

SOLUTION: This electrolyte 13 contains at least one kind selected from a first high polymer compound having a cross-linked structure wherein first compounds are cross-linked, a second compound, at least a kind out of a third compound having a heavier molar weight than the second compound and a second high polymer compound having a cross-linked structure wherein the third compounds are cross-linked, and an electrolyte salt. The first high polymer compound and the second compound form a semi-mutual penetration type high polymer network structure, the first high polymer compound and the third compound form another semi-mutual penetration type high polymer network structure, and the first high polymer compound and the second high polymer compound form a mutual penetration type high polymer network structure. High film-forming properties can be obtained by the first high polymer compound, high ion conductivity can be obtained by the second compound, and high elasticity can be obtained by the third compound or the second high polymer compound.

[What is claimed is]

[Claim 1] A composition for electrolyte comprising a 1st crosslinkable compound, a 2nd compound, and a 3rd compound which has larger molecular weight than the 2nd compound.

[Claim 2] The composition for electrolyte according to claim 1, further comprising an electrolyte salt.

[Claim 3] The composition for electrolyte according to claim 1, wherein the electrolyte salt is lithium salt.

[Claim 4] The composition for electrolyte according to claim 1, wherein the 2nd compound can form a semi-interpenetrating polymer network structure with a 1st polymer compound which is obtained by cross-linking the 1st compound, the 3rd compound can form a semi-interpenetrating polymer network structure or interpenetrating polymer network structure with the 1st polymer compound which is obtained by cross-linking the 1st compound, thereby the function which raises elasticity rather than the case of being sole.

[Claim 5] The composition for electrolyte according to claim 1, wherein the 1st compound has smaller molecular weight than said 3rd compound.

[Claim 6] The composition for electrolyte according to claim 1, wherein the 1st

compound has the unsaturated bond in which a radical polymerization can be.

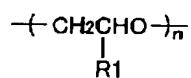
[Claim 7] The composition for electrolyte according to claim 1, wherein the 2nd compound has molecular weight being less than 10,000.

[Claim 8] The composition for electrolyte according to claim 1, wherein the 2nd compound has the ionic conductivity of  $1 \times 10^{-4}$  or more S/cm at 25°C at the time of dissolving an electrolyte salt.

[Claim 9] The composition for electrolyte according to claim 1, wherein said 2nd compound and said 3rd compound at least have ether bond, among said 1st compound, said 2nd compound, and said 3rd compound.

[Claim 10] The composition for electrolyte according to claim 1, wherein said 2nd compound and said 3rd compound has unit expressed with formula 1 among said 1st compound, said 2nd compound, and said 3rd compound.

[Formula 1]

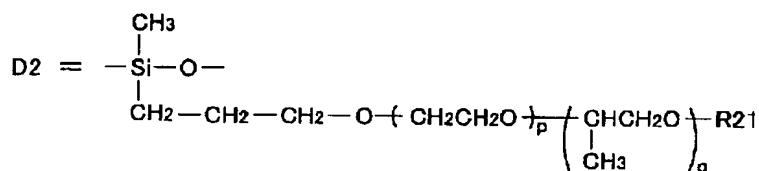
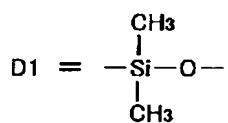
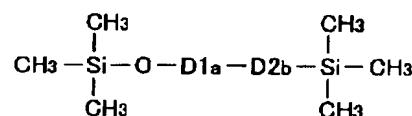


(wherein R1 expresses hydrogen, a hydrocarbon group, or group having a oxy-alkylene structure, and the repeat unit n is integers of 1 or more.)

[Claim 11] The composition for electrolyte according to claim 1, wherein said

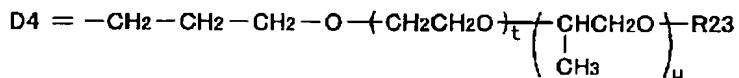
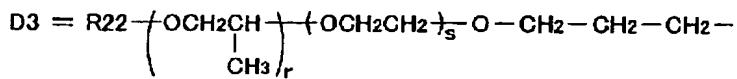
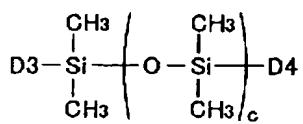
2nd compound is at least one siloxane derivative expressed with following formula 2, formula 3, or formula 4.

[Formula 2]



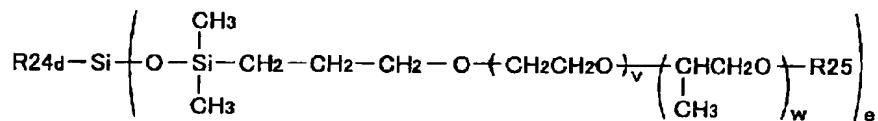
(wherein, a expresses the integer of 0 to 100, b expresses the integer of 1 to 100, p expresses the integer of 0 to 100, q expresses the integer of 0 to 100, and R21 expresses a hydrogen atom or an alkyl group. A group in which a part of hydrogen atom is substituted with halogen contains in D2 and R21.)

[Formula 3]



(wherein, c expresses the integer of 1 to 50, r, s, t, and u expresses the integer of 0 to 40, respectively, and R22 and R23 express hydrogen, alkyl group, or a substituted alkyl group in which at least one hydrogen is substituted with halogen.)

[Formula 4]



(wherein, d expresses the integer of 0 to 3, e expresses the integer of 1 to 4, d+e is surely 4 and v and w expresses the integer of 0 to 40, respectively, R24 is a methyl group and R25 expresses hydrogen, alkyl group, or a substituted alkyl group in which at least one hydrogen is substituted with halogen.)

[Claim 12] An electrolyte comprising a 1st crosslinkable compound, a 2nd compound, a 3rd compound which has larger molecular weight than the 2nd compound, and electrolyte salt.

[Claim 13] The electrolyte according to claim 12, wherein the 1st polymer compound has the structure of cross linkage by partially crosslinking with the 3rd compound.

[Claim 14] The electrolyte according to claim 12, wherein the 2nd compound can form a semi-interpenetrating polymer network structure with a 1st polymer compound which is obtained by cross-linking the 1st compound, the 3rd compound can form a semi-interpenetrating polymer network structure or interpenetrating polymer network structure with the 1st polymer compound which is obtained by cross-linking the 1st compound, thereby the function which raises elasticity rather than the case of being sole.

[Claim 15] The electrolyte according to claim 12, wherein the 1st compound has smaller molecular weight than said 3rd compound.

[Claim 16] The electrolyte according to claim 12, wherein the 1st compound has the unsaturated bond in which a radical polymerization can be.

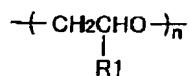
[Claim 17] The electrolyte according to claim 12, wherein the 2nd compound has molecular weight being less than 10,000.

[Claim 18] The electrolyte according to claim 12, wherein the 2nd compound has the ionic conductivity of  $1 \times 10^{-4}$  or more S/cm at 25°C at the time of dissolving an electrolyte salt.

[Claim 19] The electrolyte according to claim 12, wherein said 2nd compound and said 3rd compound at least have ether bond, among said 1st compound, said 2nd compound, and said 3rd.

[Claim 20] The electrolyte according to claim 12, wherein said 2nd compound and said 3rd compound has unit expressed with formula 5 among said 1st compound, said 2nd compound, and said 3rd compound.

[Formula 5]

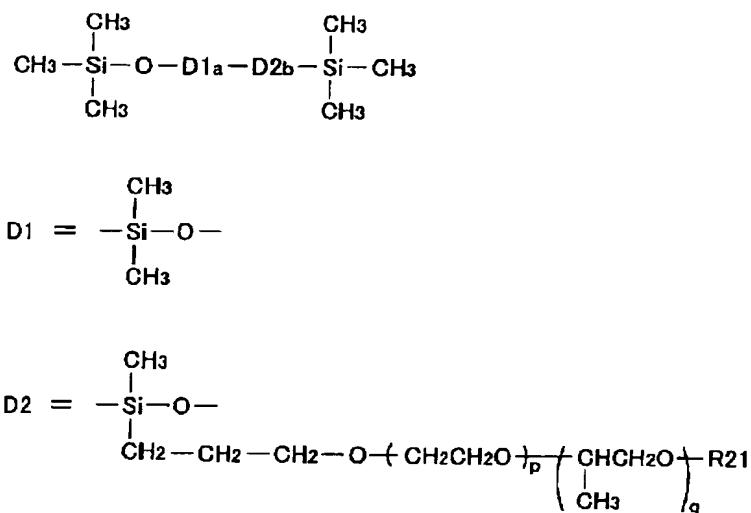


(wherein R1 expresses hydrogen, a hydrocarbon group, or group having a oxy-alkylene structure, and the repeat unit n is integers of 1 or more.) [Claim 21]

The electrolyte according to claim 12, wherein said 2nd compound is at least

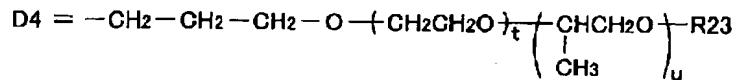
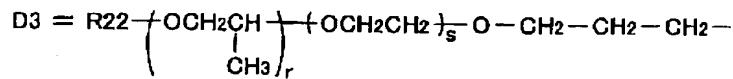
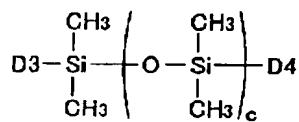
one siloxane derivative expressed with following formula 6, formula 7, or formula 8.

[Formula 6]



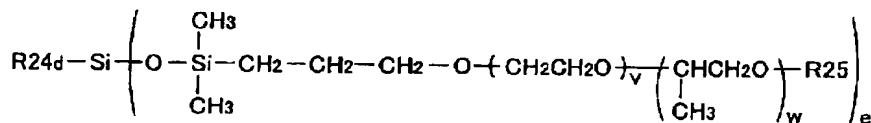
(wherein, a expresses the integer of 0 to 100, b expresses the integer of 1 to 100, p expresses the integer of 0 to 100, q expresses the integer of 0 to 100, and R21 expresses a hydrogen atom or an alkyl group. A group in which a part of hydrogen atom is substituted with halogen contains in D2 and R21.)

[Formula 7]



(wherein, c expresses the integer of 1 to 50, r, s, t, and u expresses the integer of 0 to 40, respectively, and R22 and R23 express hydrogen, alkyl group, or a substituted alkyl group in which at least one hydrogen is substituted with halogen.)

[Formula 8]



(wherein, d expresses the integer of 0 to 3, e expresses the integer of 1 to 4, d+e is surely 4 and v and w expresses the integer of 0 to 40, respectively, R24 is a methyl group and R25 expresses hydrogen, alkyl group, or a substituted alkyl group in which at least one hydrogen is substituted with halogen.)

[Claim 22] The electrolyte according to claim 12, wherein the electrolyte salt is lithium salt.

[Claim 23] A method of preparing electrolyte, comprising cross-linking a 1st compound in a mixture of a crosslinkable 1st compound, a 2nd compound, a 3rd compound which has larger molecular weight than the 2nd compound.

[Claim 24] A method of preparing electrolyte, comprising adding an electrolyte salt before cross-linking a 1st compound in a mixture of a crosslinkable 1st compound, a 2nd compound, a 3rd compound which has larger molecular weight than the 2nd compound.

[Claim 25] A battery comprising a positive electrode, a negative electrode, and an electrolyte, the electrolyte comprising a 1st crosslinkable compound, a 2nd compound, a 3rd compound which has larger molecular weight than the 2nd compound, and electrolyte salt.

[Claim 26] A battery according to claim 25, wherein the 1st polymer compound has the structure of cross linkage by partially crosslinking with the 3rd compound.

[Claim 27] A battery according to claim 25, wherein the 2nd compound can form a semi-interpenetrating polymer network structure with a 1st polymer compound

which is obtained by cross-linking the 1st compound, the 3rd compound can form a semi-interpenetrating polymer network structure or interpenetrating polymer network structure with the 1st polymer compound which is obtained by cross-linking the 1st compound, thereby the function which raises elasticity rather than the case of being sole.

[Claim 28] A battery according to claim 25, wherein the 1st compound has smaller molecular weight than said 3rd compound.

[Claim 29] A battery according to claim 25, wherein the 1st compound has the unsaturated bond in which a radical polymerization can be.

[Claim 30] A battery according to claim 25, wherein the 2nd compound has molecular weight being less than 10,000.

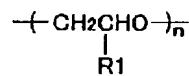
[Claim 31] A battery according to claim 25, wherein the 2nd compound has the ionic conductivity of  $1 \times 10^{-4}$  or more S/cm at 25°C at the time of dissolving an electrolyte salt.

[Claim 32] A battery according to claim 25, wherein said 2nd compound and said 3rd compound at least have ether bond, among said d 1st compound, said 2nd compound, and said 3rd.

[Claim 33] A battery according to claim 25, wherein said 2nd compound and

said 3rd compound has unit expressed with formula 9 among said 1st compound, said 2nd compound, and said 3rd compound, and said 3rd compound.

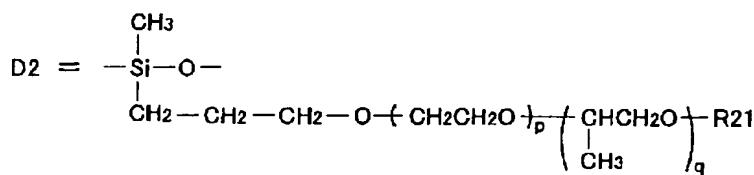
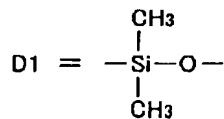
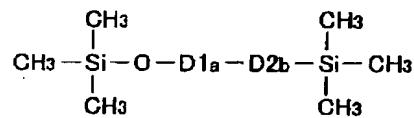
[Formula 9]



(wherein R1 expresses hydrogen, a hydrocarbon group, or group having a oxy-alkylene structure, and the repeat unit n is integers of 1 or more.) [Claim 34]

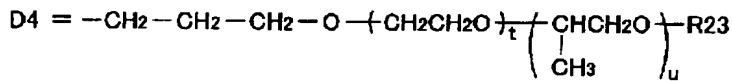
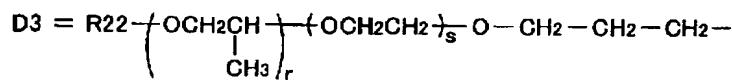
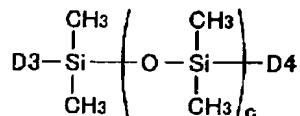
The battery according to claim 25, wherein said 2nd compound is at least one siloxane derivative expressed with following formula 10, formula 11, or formula 12.

[Formula 10]



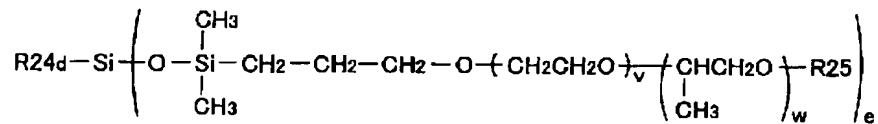
(wherein, a expresses the integer of 0 to 100, b expresses the integer of 1 to 100, p expresses the integer of 0 to 100, q expresses the integer of 0 to 100, and R21 expresses a hydrogen atom or an alkyl group. A group in which a part of hydrogen atom is substituted with halogen contains in D2 and R21.)

[Formula 11]



(wherein, c expresses the integer of 1 to 50, r, s, t, and u expresses the integer of 0 to 40, respectively, and R22 and R23 express hydrogen, alkyl group, or a substituted alkyl group in which at least one hydrogen is substituted with halogen.)

[Formula 12]



(wherein, d expresses the integer of 0 to 3, e expresses the integer of 1 to 4, d+e is surely 4 and v and w expresses the integer of 0 to 40, respectively, R24 is a methyl group and R25 expresses hydrogen, alkyl group, or a substituted alkyl group in which at least one hydrogen is substituted with halogen.

[Claim 35] The battery according g to claim 25, wherein the electrolyte salt is lithium salt.

[Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention relates to the electrolyte containing a high molecular compound and an electrolyte salt, its manufacture approach and its constituent for electrolytes, and the cell that used the electrolyte for the list.

[0002]

[Description of the Prior Art] In recent years, portable electronic devices, such as a camcorder/movie (video tape recorder), a cellular phone, or a laptop computer, are spreading quickly, and the further high performance is called for about the electrochemistry device carried in these electronic equipment, for example, a rechargeable battery.

[0003] Conventionally, as a medium which manages ionic conduction in a rechargeable battery, the liquefied electrolyte made to dissolve an electrolyte salt in water or an inflammable organic solvent has been used. However, a liquefied electrolyte has problems, such as a liquid spill, and, for this reason, needed to secure confidentiality using the metal container. Therefore, this kind of cell was heavy and its complicatedness was low in connection with the sealing process. Then, the research using the so-called solid electrolyte which

consists of an ion conductivity solid-state is made actively. There is an advantage, like the degree of freedom of shape selectivity is high by the film moldability which there are no worries about a liquid spill, and could attain lightweight and simplification of a sealing process, and excelled in the polymer.

[0004] This solid electrolyte consists of for example, a matrix macromolecule and a possible electrolyte salt of ionic dissociation. Among these, the matrix macromolecule holds ionic dissociation ability and is bearing the role which solidifies this ion conductivity solid-state, and a role of a solvent to an electrolyte salt. Such a solid electrolyte is reported in 1978 to have obtained the ionic conductivity of  $1 \times 10^{-7}$  S/cm extent by Armand and others of the Grenoble university (France) by the system which dissolved lithium perchlorate in polyethylene oxide, and examination has been performed still more actively current since then about the various polymeric materials focusing on the polymer which has polyether association.

[0005]

[Problem(s) to be Solved by the Invention] However, in the individual electrolyte which consists of a high molecular compound and an electrolyte salt, since ion conducts using the local thermal motion of a macromolecule, i.e., segmental

motion, compared with the viscous low liquefied electrolyte, ionic conductivity is low and has not resulted in utilization. Although complicated molecular designs, such as exclusion of a crystal part which checks the segmental motion of a macromolecule, or reduction in glass transition temperature, were needed as an approach of raising the ionic conductivity of an individual electrolyte, such an amorphous polymer had generally the problem that ionic conductivity was not enough, either in the reinforcement as film deficiently.

[0006] This invention was made in view of this trouble, and that purpose is in offering the cell using the constituent for electrolytes in which high membrane formation nature and high ionic conductivity can be shown, an electrolyte, its manufacture approach, and this electrolyte.

[0007]

[Means for Solving the Problem] The constituent for electrolytes by this invention contains the 1st compound which can construct a bridge, the 2nd compound, and the 3rd compound which has bigger molecular weight than this 2nd compound.

[0008] The electrolyte by this invention contains at least one sort in the 2nd high molecular compound which has the structure of cross linkage by which the

bridge was constructed over the 1st high molecular compound which has the structure of cross linkage by which the bridge was constructed over the 1st compound, the 2nd compound, the 3rd compound which has larger molecular weight than this 2nd compound, and this 3rd compound, and an electrolyte salt.

[0009] The manufacture approach of the 1st electrolyte by this invention includes the process over which the 1st compound is made to construct a bridge about the 1st compound which can construct a bridge, the 2nd compound, the 3rd compound which has bigger molecular weight than the 2nd compound, and mixture with an electrolyte salt.

[0010] About the mixture of the 1st compound which can construct a bridge, the 2nd compound, and the 3rd compound which has bigger molecular weight than the 2nd compound, the manufacture approach of the 2nd electrolyte by this invention includes the process which adds an electrolyte salt, after making the 1st compound construct a bridge.

[0011] The cell by this invention is equipped with an electrolyte with a positive electrode and a negative electrode, and an electrolyte contains at least one sort in the 2nd high molecular compound which has the structure of cross linkage by which the bridge was constructed over the 1st high molecular compound which

has the structure of cross linkage by which the bridge was constructed over the 1st compound, the 2nd compound, the 3rd compound which has bigger molecular weight than this 2nd compound, and this 3rd compound, and an electrolyte salt.

[0012] In the constituent for electrolytes by this invention, since the 1st compound which can construct a bridge, the 2nd compound, and the 3rd compound which has bigger molecular weight than this 2nd compound are included, the electrolyte in which high membrane formation nature, high ion conductivity, and high elasticity are shown is obtained by making the 1st compound construct a bridge.

[0013] In the electrolyte by this invention, the 2nd compound, the 3rd compound, the 2nd high molecular compound, and an electrolyte salt exist between the 1st high molecular compound which has the structure of cross linkage by which the bridge was constructed over the 1st compound. Among these, improvement in membrane formation nature is achieved with the 1st high molecular compound, ion conductivity improvement is achieved with the 2nd compound, and improvement in elasticity is achieved with the 3rd compound and the 2nd high molecular compound.

[0014] By the manufacture approach of the 1st electrolyte by this invention, a bridge is constructed over the 1st compound about the mixture of the 1st compound, the 2nd compound, the 3rd compound, and an electrolyte salt.

[0015] By the manufacture approach of the 2nd electrolyte by this invention, about the mixture of the 1st compound, the 2nd compound, and the 3rd compound, after a bridge is constructed over the 1st compound, an electrolyte salt is added.

[0016] By the cell by this invention, when the ion produced by dissociation of an electrolyte salt moves between a negative electrode and a positive electrode in the inside of an electrolyte, it discharges. Here, since it has the electrolyte of this invention, it has high membrane formation nature, high ion conductivity, and high elasticity, and the outstanding cell engine performance is shown.

[0017]

[Embodiment of the Invention] Hereafter, the gestalt of operation of this invention is explained to a detail.

[0018] The electrolyte concerning the gestalt of 1 operation of this invention contains at least one sort in the 2nd high molecular compound which has the structure of cross linkage by which the bridge was constructed over the 1st high

molecular compound which has the structure of cross linkage by which the bridge was constructed over the 1st compound, the 2nd compound, the 3rd compound which has bigger molecular weight than the 2nd compound, and this 3rd compound, and an electrolyte salt. As for the 2nd compound, what does not dissolve an electrolyte salt and does not have the functional group which can construct a bridge is desirable, and, as for the 1st high molecular compound and the 2nd compound, it is desirable to form both the half-invasion mold macromolecule network structure (semi-IPN;interpenetrating polymer network).

[0019] Even if the 3rd compound has the functional group which can construct a bridge, it is not necessary to have it, and as for the 1st high molecular compound and the 3rd compound, it is desirable to form the mutual invasion mold macromolecule network structure (IPN) or both the half-invasion mold macromolecule network structure (semi-IPN). For example, when it has the functional group over which the 3rd compound can construct a bridge, as for the 2nd high molecular compound, it is desirable to form the 1st high molecular compound and the mutual invasion mold macromolecule network structure, and to form the 2nd compound and both the half-invasion mold macromolecule

network structure. In this case, the structure of cross linkage by which the 1st compound and the 3rd compound constructed the bridge in the part, and the bridge was constructed over the 3rd compound by a part of 1st high molecular compound may exist. When it does not have the functional group over which the 3rd compound can construct a bridge, as for the 3rd compound, it is desirable to form the 1st high molecular compound and both the half-invasion mold macromolecule network structure.

[0020] In addition, the condition of dissolving without, as for saying the semi-interpenetrating polymer network structure, one side's joining together chemically, and not combining another side but joining together chemically mutually is said. The mutual invasion mold macromolecule network structure says the condition with which that over which mix, both sides made what has a combinable functional group chemically construct a bridge over, and each constructed the bridge became entangled, or the condition that dissolve mutually and both sides are constructing the bridge partially.

[0021] Generally, since the bridge is constructed over the 1st high molecular compound, it has membrane formation nature and strong film reinforcement, but since molecular motion is controlled by bridge formation and the mobility of

dissolved ion becomes small, ionic conductivity is small. On the other hand, since a bridge is not constructed, the 2nd compound has a high momentum of a molecule, and by forming the 1st high molecular compound and both the half-invasion mold macromolecule network structure, crystallinity also falls and it shows high ionic conductivity in a large temperature requirement. So, with the gestalt of this operation, high membrane formation nature and high ionic conductivity can be obtained now by including the 1st high molecular compound and the 2nd compound, respectively.

[0022] Moreover, although fixed film reinforcement is obtained with the 1st high molecular compound only with the 1st high molecular compound and the 2nd compound, elasticity is low, and the application to a flexible device etc. is difficult. So, with the gestalt of this operation, including at least one sort in the 3rd compound and the 2nd high molecular compound, when they and the 1st high molecular compound form both the semi-interpenetrating polymer network structure or the interpenetrating polymer network structure, elasticity can be raised rather than the case of being independent.

[0023] Therefore, as the 1st polymer compound, it is preferred crosslinking density being large since film strength is more strong, for example, the one

smaller than the 3rd compound of the molecular weight of the 1st compound is desirable.

[0024] as the 2nd compound, it is preferred viscosity being low since ionic conductivity is more high, for example the smaller one of molecular weight is desirable. A liquefied thing is specifically desirable in ordinary temperature, and, as for the weight average molecular weight of the 2nd compound, it is desirable that it is less than 10,000. However, if the molecular weight of the 2nd compound is too small conversely, since a solution retention problem will arise from relation, such as vapor pressure, it is more desirable than low-boiling point solvents, such as propylene carbonate, that it is extent which can maintain low vapor pressure, for example. In addition, as for the ionic conductivity in 25°C at the time of dissolving the electrolyte salt of the 2nd compound, it is desirable that they are  $1 \times 10^{-4}$  or more S/cm.

[0025] As the 3rd compound, it is preferred molecular weight being large since elasticity is high for example, it is more desirable if weight average molecular weight is 10,000 or more and further 100,000 or more. However, while the thing of the crystalline shape of amorphous small can make elasticity high as the 3rd compound, since ionic conductivity can be made high, it is desirable.

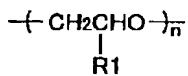
Moreover, it is more desirable for the 3rd compound to have been desirable since the direction which has the functional group which can construct a bridge strengthened film reinforcement more, namely, to contain the 2nd high molecular compound. Furthermore, since the direction which has the high free end side chain of molecular motion nature can make ionic conductivity high, the 3rd compound is desirable.

[0026] The one fewer than the 2nd compound of the content of the 3rd compound and the 2nd high molecular compound is desirable. It is because the 2nd compound bears the great portion of ionic conduction, so ionic conductivity will become low if there are many contents of the 3rd compound and the 2nd high molecular compound.

[0027] In addition, the 2nd compound and the 3rd compound have the desirable thing of the 1st high molecular compound, the 2nd compound, and the 3rd compound for which it has ether linkage, respectively, for example, it is desirable to include the configuration unit expressed with formula 13 at least. To contain is more more desirable although these do not need to be included about the 1st high molecular compound. While raising compatibility, it is because ionic conductivity can be raised more.

[0028]

[Formula 13]



(wherein R1 expresses hydrogen, a hydrocarbon group, or group having a oxy-alkylene structure, and the repeat unit n is integers of 1 or more.) [0029] Among these, as the 1st high molecular compound, what has the structure where the bridge was constructed over either at least the straight chain-like 1st compound or, and the 1st compound of a pectinate form in the bridge formation radical is mentioned, and you may have the side chain, for example. In addition, the 1st high molecular compound may have annular atomic arrangement in some of principal chains or side chains that what is necessary is just to include chain-like atomic arrangement in part at least, for example. Moreover, the bridge may be constructed over what it has at one end even if the bridge is constructed over what has the functional group which can construct a bridge over both ends that what is necessary is just to construct a bridge over the 1st compound which has at least one functional group over which the 1st high molecular compound can construct a bridge, if the 1st compound is a straight chain-like. The bridge may

be constructed over what has the functional group which can construct a bridge over at least one end as long as the 1st compound is a pectinate form. Namely, the 1st high molecular compound should just form the three-dimensional network which can hold the 2nd compound and an electrolyte salt.

[0030] However, in order to realize higher ionic conductivity, it is desirable to have the structure where the bridge was constructed over the 1st compound which has at least one free end which the functional group which can construct a bridge is not allotted and includes ether linkage. It is because it will have the free end side chain which branching structure is introduced into a three-dimensional network by this, and includes ether linkage. for example, the thing which has the functional group which can construct a bridge over both ends if the 1st compound is a straight chain-like, in addition, it is desirable that the bridge is constructed over what has the functional group which can construct a bridge over one end. Moreover, if the 1st compound is a pectinate form, it is desirable that the bridge is constructed over what has at least one free end to which the functional group which can construct a bridge is not allotted. Furthermore, when the bridge is constructed over the straight chain-like 1st compound, and the 1st compound of a pectinate form, it is desirable to have at

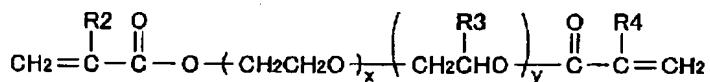
least one of these.

[0031] The bridge could be constructed over the thing by which the bridge was constructed over the 1st compound which has the unsaturated bond in which radical polymerizations, such as a functional group which association like a throat, such as ether linkage, an ester bond, or a urethane bond, is sufficient as the structure of cross linkage of the 1st high molecular compound, for example, has an allyl group, a vinyl group, an acrylate radical, a methacrylate radical, or cycloolefin structure, are possible in them, or the 1st compound which has an active hydrogen radical in them. Moreover, the structure of cross linkage from which plurality differs may exist in 1 compound.

[0032] What has the structure where what specifically has the structure of cross linkage by which the bridge was constructed over the ester compound as the 1st high molecular compound was mentioned, and the bridge was constructed over at least one sort in a monoester compound, a diester compound, and a triester compound especially is desirable. What has the structure where the bridge was constructed over the diester compound including the oxy-alkylene structure expressed with the general formula shown in formula 14 as an example is mentioned.

[0033]

[Formula 14]

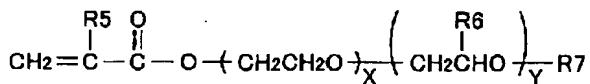


R2, R3, and R4 express a hydrogen atom or the alkyl group of carbon numbers 1-6 among a formula, and even if each is mutually the same, it may differ. x and y may be the integers of  $x \geq 1$  and  $y \geq 0$  or  $x \geq 0$ , and  $y \geq 1$ , and a value which is mutually different also with the same value is sufficient as them.

[0034] In this case, it is more more desirable to have the structure where the bridge was constructed over the monoester compound including the oxyalkylene structure expressed with the general formula shown in formula 15 in addition to the structure where the bridge was constructed over the diester compound expressed with the general formula shown in formula 14, for example. It is because a free end side chain including ether linkage is introduced and higher ionic conductivity can be obtained, as mentioned above.

[0035]

[Formula 15]



R5, R6, and R7 express a hydrogen atom or the alkyl group of carbon numbers 1-6 among a formula, and even if each is mutually the same, it may differ. X and Y may be the integers of  $X \geq 1$  and  $Y \geq 0$  or  $X \geq 0$ , and  $Y \geq 1$ , and a value which is mutually different also with the same value is sufficient as them.

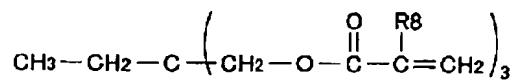
[0036] As for the structure where the bridge was constructed over the diester compound, and the structure where the bridge was constructed over the monoester compound, it is desirable in that case that it is within the limits of 5.0 or less more greatly than 0 at the weight ratio (a monoester compound / diester compound) of a monoester compound to a diester compound. It is because a three-dimensional network cannot be constituted if there is little structure where the bridge was constructed over the diester compound, but the free end side chain which includes ether linkage when there is little structure where the bridge was constructed over the monoester compound decreases and ionic conductivity cannot be made high.

[0037] Moreover, what has the structure where the bridge was constructed over the triester compound including the oxy-alkylene structure expressed with the general formula shown in formula 16 or formula 17 as other examples is

mentioned.

[0038]

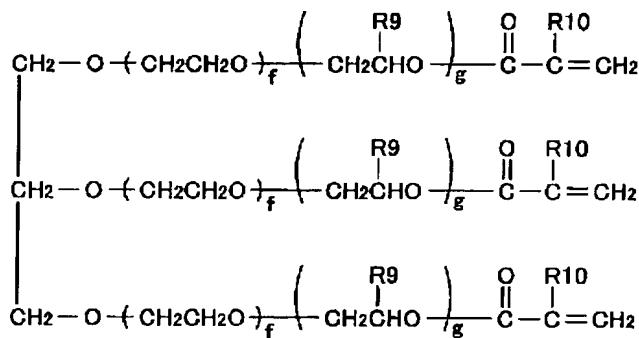
[Formula 16]



R8 expresses a hydrogen atom or the alkyl group of carbon numbers 1-6 among a formula.

[0039]

[Formula 17]



Among the formula, R9 and R10 express a hydrogen atom or the alkyl group of carbon numbers 1-6, and even if mutually the same, they may differ. f and g may be the integers of  $f \geq 1$  and  $g \geq 0$  or  $f \geq 0$ , and  $g \geq 1$ , and a value which is different also with the same value is sufficient as them.

[0040] It is more desirable to have the structure where the bridge was constructed over the monoester compound expressed with the general formula shown in formula 15 in addition to the structure where the bridge was constructed over the triester compound expressed with the general formula shown in formula 16 and formula 17, as mentioned above also in this case.

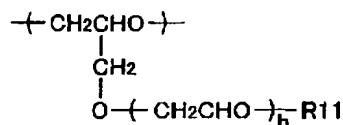
[0041] In addition, although especially the presentation ratio of f and g of the triester compound shown in formula 17 is not limited, it is desirable that it is within the limits of  $0.1 \leq g/f \leq 4$ . It is because it will become weak if many [ if there are too few oxyethylene units, a mechanical strength will fall, and / too ]. Moreover, although any of the shape of the letter of a block or random are sufficient as the joint format of an oxyethylene unit and an oxy-alkylene unit, the shape of random is more desirable.

[0042] What has the structure where the bridge was constructed in the functional group over which the copolymer which includes the configuration unit shown in the configuration unit shown in formula 18 and formula 19 as the 1st high molecular compound can construct a bridge is mentioned. According to this, since the free end side chain which includes ether linkage by the configuration unit shown in formula 18 is introduced and high ionic conductivity

can be obtained, it is desirable.

[0043]

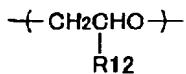
[Formula 18]



R11 expresses at least one sort in the alkyl group of carbon numbers 1-12, the alkenyl radical of carbon numbers 2-18, the cycloalkyl radical of carbon numbers 3-8, the aryl group of carbon numbers 6-18, the aralkyl radical of carbon numbers 7-12, and a tetrahydropyranyl group among a formula, and h is the integer of  $1 \leq h \leq 12$ . In addition, even if R11 and h are the same in all configuration units, they may be good, and they may change with configuration units.

[0044]

[Formula 19]



R12 expresses among a formula at least one sort in at least one side of the

functional groups which have at least one side of the functional groups which have an active hydrogen radical and an unsaturated bond or an active hydrogen radical, and an unsaturated bond, a hydrogen atom, an alkyl group, an alkenyl radical, a cycloalkyl radical, and an aryl group. In addition, even if R12 is the same in all configuration units, it may be good, and it may change with configuration units.

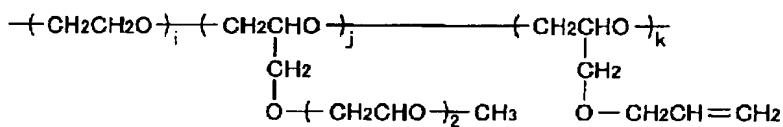
[0045] Any of the shape of the letter of a block or random are sufficient as the joint format with the configuration unit shown in the configuration unit which especially the presentation ratio with the configuration unit shown in the configuration unit shown in formula 18 in this copolymer and formula 19 is not limited, and was shown in formula 18, and formula 19. Incidentally, in this copolymer, the functional group which can construct a bridge is a functional group which has an active hydrogen radical and an unsaturated bond. The functional group which has the allyl group in which a radical polymerization is possible, a vinyl group, an acrylate radical, a methacrylate radical, or cycloolefin structure as a functional group which has an unsaturated bond is mentioned.

[0046] That by which the bridge was constructed over the copolymer specifically expressed with the general formula shown in formula 20 as the 1st high

molecular compound which has such structure is desirable. h [ in / in the copolymer shown in formula 20 / formula 18 ] includes the configuration unit which R11 becomes from a methyl group, the configuration unit which R12 in formula 19 becomes from a hydrogen atom, and the configuration unit which R12 in formula 19 becomes from an allyl group by two.

[0047]

[Formula 20]



Among a formula, i, j, and k may be one or more integers, and a value which is mutually different also with the same value is sufficient as them.

[0048] In addition, two or more compounds with which such structures differ may be mixed and used for the 1st high molecular compound. For example, the compound which has the structure where the bridge was constructed over the copolymer including the compound which has the structure where the bridge was constructed over the ester compound shown in formula 14, and the configuration unit shown in the configuration unit shown in formula 18 and

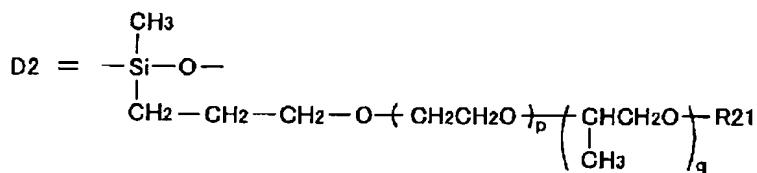
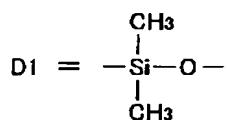
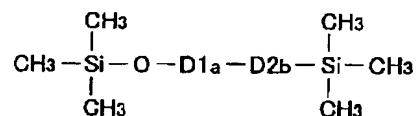
formula 19 may be mixed and used. Moreover, the thing over which two or more 1st compounds with which structures differ were made to construct a bridge may be used.

[0049] It is n ( $\text{SiOCH}_3\text{R}$ ) which has the chain combination of silicon (Si) and oxygen (O) in a basic frame and by which the side chain radical R was added to silicon as the 2nd compound, for example. The shape type siloxane derivative of a chain expressed is mentioned. In this case, a substituent or a side chain radical R is an organic radical of monovalence, and serves as the structure where an electrolyte salt can be dissolved, by choosing a side chain radical R moderately. Moreover, as for this siloxane derivative, it is desirable to include ether linkage in a substituent or a side chain radical R, as mentioned above. As such a side chain radical R, there are alkoxy groups, such as an ethoxy radical, a propoxy group, a butoxy radical, and a methoxy group. Moreover, the hydrogen in a side chain radical R may be replaced by halogens, such as boron or a fluorine.

[0050] Specifically as such a siloxane derivative, it is desirable that what is expressed with the general formula shown in formula 21, formula 22, or formula 23 is mentioned, and at least one of sorts of these is included.

[0051]

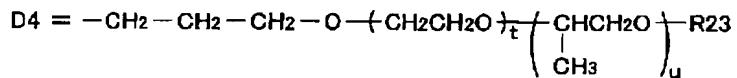
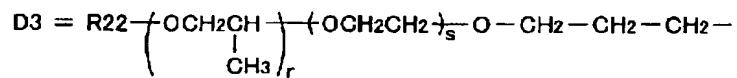
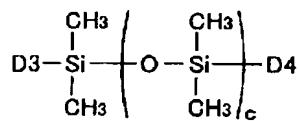
[Formula 21]



(wherein, a expresses the integer of 0 to 100, b expresses the integer of 1 to 100, p expresses the integer of 0 to 100, q expresses the integer of 0 to 100, and R21 expresses a hydrogen atom or an alkyl group. A group in which a part of hydrogen atom is substituted with halogen contains in D2 and R21.) At the time of  $b > 1$ , even if b D2 is the same, it may differ. A value which is different also with the same value is sufficient as p and q.

[0052]

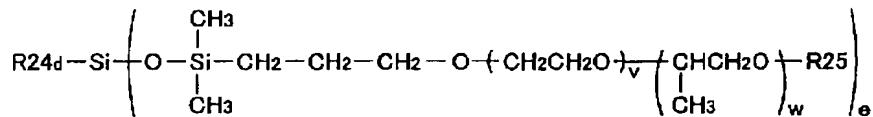
[Formula 22]



(wherein, c expresses the integer of 1 to 50, r, s, t, and u expresses the integer of 0 to 40, respectively, and R<sub>22</sub> and R<sub>23</sub> express hydrogen, alkyl group, or a substituted alkyl group in which at least one hydrogen is substituted with halogen. A value which is different also with the same value is sufficient as r, s, and t and u, and even if R<sub>22</sub> and R<sub>23</sub> are the same, they may differ.

[0053]

[Formula 23]



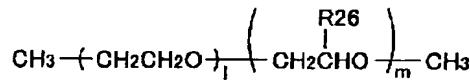
(wherein, d expresses the integer of 0 to 3, e expresses the integer of 1 to 4, d+e is surely 4 and v and w expresses the integer of 0 to 40, respectively, R<sub>24</sub>

is a methyl group and R25 expresses hydrogen, alkyl group, or a substituted alkyl group in which at least one hydrogen is substituted with halogen.) A value which is different also with the same value is sufficient as v and w.

[0054] The polyalkylene glycol wood ether expressed with the general formula shown in formula 24 as the 2nd compound is mentioned. Two or more sorts of compounds may be mixed and used, for example, it may mix with polyalkylene glycol wood ether and a siloxane derivative to the 2nd compound, and you may use for it.

[0055]

[Formula 24]



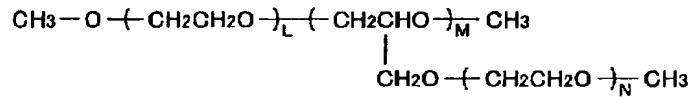
Among a formula, l and m express mol % in the molecule of each unit, respectively, and fulfill the conditions of  $l+m=100$  and  $0 < l \leq 100$ . R26 expresses the radical containing the radical or oxy-alkylene unit by which a part of hydrogen atom [at least] contained in a hydrogen atom, an alkyl group, and an alkyl group was permuted by the halogen atom.

[0056] As the 3rd compound, the polyether derivative which has ether structure,

or the poly FOSUFAZEN derivative is mentioned to a polyether and a side chain, for example. Polyethylene oxide, the polyethylene oxide derivative expressed with the general formula shown in formula 25, or the poly FOSUFAZEN derivative expressed with the general formula shown in formula 26 is more specifically mentioned. Two or more sorts of these compounds may be mixed and used for the 3rd compound and the 2nd high molecular compound.

[0057]

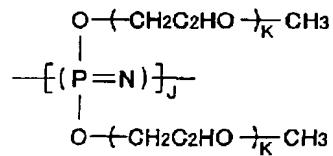
[Formula 25]



L and M express the mole ratio in the molecule of each unit among a formula, respectively, and N is the integer of 1 to 4.

[0058]

[Formula 26]



One or more integers and K of J are the integers of 1 to 8 among a formula.

[0059] An electrolyte salt is for giving ion conductivity by dissociation, and is expressed with the general formula shown in formula 27.

[Formula 27] A expresses a cation among AB type and B expresses an anion.

[0060] A light metal salt etc. is mentioned as this electrolyte salt, and, specifically, it is used according to the purpose from alkaline-earth-metals salts, such as alkali-metal salts, such as a lithium (Li) salt, a sodium (Na) salt, or a potassium (K) salt, a calcium (calcium) salt, or a magnesium (Mg) salt, etc. In addition, the examples of the lithium salt are LiClO<sub>4</sub>, LiBF<sub>4</sub>, LiPF<sub>6</sub>, LiAsF<sub>6</sub>, LiAlCl<sub>4</sub>, LiSbF<sub>6</sub>, LiCF<sub>3</sub>SO<sub>3</sub>, LiN(CF<sub>3</sub>SO<sub>2</sub>)<sub>2</sub>, LiC<sub>4</sub>F<sub>9</sub>SO<sub>3</sub>, LiCF<sub>3</sub>CO<sub>2</sub>, and LiN(CF<sub>3</sub>CO<sub>2</sub>)<sub>2</sub>, the examples of sodium salt are NaClO<sub>4</sub>, NaBF<sub>4</sub>, NaSCN, etc, and the examples of potassium salt are KBF<sub>4</sub> etc. The need is accepted, a gap, one sort, or two sorts or more are mixed, and these are used.

[0061] The electrolyte which has such a configuration can be manufactured as follows using the following constituents for electrolytes.

[0062] First, the constituent for electrolytes containing the 1st compound which has the functional group which can construct a bridge, the 2nd compound, the 3rd compound, and an electrolyte salt is prepared. It is as having mentioned

above about these 1st compounds, the 2nd compound, the 3rd compound, and an electrolyte salt. In addition, the triester compound shown in the diester compound shown in formula 14, the monoester compound shown in formula 15, formula 16, or formula 17 as an example of the 1st compound, a copolymer including the configuration unit shown in the configuration unit shown in formula 18 and formula 19, etc. are mentioned.

[0063] Subsequently, these 1st compounds, the 2nd compound, the 3rd compound, and an electrolyte salt are mixed, and uniform mixture is produced. A polymerization initiator is also added if needed in that case. A polymerization initiator is needed except when the exposure of an electron ray performs a polymerization, and it has a photopolymerization initiator and a thermal polymerization initiator.

[0064] As a photopolymerization initiator, there is acetophenone, TORIKURORO acetophenone, 2-hydroxy-2-methylpropiophenone, 2-hydroxy-2-methyl iso propiophenone, 1-hydroxy cyclohexyl ketone, BENZO iso ether, 2, and 2-diethoxy acetophenone or benzyl dimethyl ketal, for example. Moreover, as a thermal polymerization initiator, there is initiators, such as elevated-temperature polymerization initiators, such as cumene hydroperoxide, t-butyl

hydroperoxide, JIKUMIRU peroxide, or G t-butyl peroxide, or a benzoyl peroxide, lauroyl peroxide, persulfate, or azobisisobutyronitril, or a redox initiator, for example. As a polymerization initiator, 1 of sorts of these and two sorts or more may be mixed and used.

[0065] As for the addition of a polymerization initiator, it is desirable to consider as within the limits of the 0.1 weight section - 1.0 weight section for example, to the 1st compound 100 weight section. It is because effectiveness will not change even if a rate of polymerization falls remarkably and adds exceeding the 1.0 weight sections if fewer than the 0.1 weight section. In addition, when it has the functional group over which the 3rd compound can construct a bridge, it is desirable to add further the polymerization initiator of the 0.1 weight section - 1.0 weight section within the limits for example, to the 3rd compound 100 weight section.

[0066] Then, ultraviolet rays, an electron ray, X-rays, a gamma ray, microwave, or a RF is irradiated at this mixture, or this mixture is heated, and the polymerization of the 1st compound is carried out. In having the functional group over which the 3rd compound can construct a bridge in that case, it carries out the polymerization also of the 3rd compound to coincidence.

Although especially the conditions of a polymerization reaction are not limited, a polymerization reaction is considered from the simple nature of equipment, and a cost side, and UV irradiation or its heating polymerization is desirable. Thereby, while a bridge is constructed over the 1st compound, a bridge is constructed over the 3rd compound if needed, and the electrolyte concerning the gestalt of this operation is obtained.

[0067] In addition, in case the 1st compound, the 2nd compound, the 3rd compound, and an electrolyte salt are mixed, proper organic solvents, such as an acetonitrile, are used and you may make it form mixture. In this case, desiccation processing is performed, and after removing an organic solvent, you may make it make the 1st compound construct a bridge, after performing desiccation processing of leaving or heating in air and under reduced pressure after making the 1st compound construct a bridge, and making it remove an organic solvent and obtaining that mixture.

[0068] Moreover, when, producing the electrolyte used for the lithium cell using alkali-metal ion, a lithium ion battery, and a sodium cell for example, as for the solvent and polymerization initiator at the time of mixing the constituent for electrolytes, and these, it is desirable to fully perform dehydration processing,

and it is desirable to also make the ambient atmosphere under manufacture into low humidity. This is because there are some which are decomposed by reacting with moisture depending on the class of alkali-metal salt. Moreover, it is because alkali metal and moisture react violently in a negative electrode when it uses as a cell.

[0069] Moreover, this electrolyte can be manufactured even if as follows.

[0070] First, the constituent for electrolytes containing the 1st compound which has the functional group which can construct a bridge, the 2nd compound, and the 3rd compound is prepared. Subsequently, the 1st compound, the 2nd compound, and the 3rd compound are mixed. A polymerization initiator is mixed if needed in that case. Then, the polymerization of the 3rd compound is carried out the 1st compound and if needed like the previous manufacture approach, and the mixture containing at least one sort of the 1st high molecular compound, the 2nd compound, the 3rd compound, and the 2nd macromolecule is formed. Dissolve an electrolyte salt in an organic solvent after it, this mixture is made to permeate, and desiccation removal of the organic solvent is carried out. Thereby, the electrolyte concerning the gestalt of this operation is obtained.

[0071] Thus, the electrolyte manufactured is used for a cell as follows. Here, the example of the rechargeable battery which used the lithium is given and it explains below with reference to a drawing.

[0072] FIG. 1 expresses the cross-section structure of the rechargeable battery using the electrolyte concerning the gestalt of this operation. In addition, what was shown in FIG. 1 is called so-called paper mold. The laminating of this rechargeable battery is carried out through the electrolyte 13 which requires a negative electrode 11 and a positive electrode 12 for the gestalt of this operation. The insulating packing 14 is arranged in these side faces, and it is sealed by pasting up with the periphery section of the sheathing members 15 and 16 of the pair arranged so that a negative electrode 11 and a positive electrode 12 might be inserted in between.

[0073] The negative electrode 11 contains a material into which lithium is intercalated or deintercalated, or lithium metal as a negative active material, and contains binders, such as polyvinylidene fluoride, further if needed.

[0074] Considering the material into which lithium is intercalated or deintercalated, carbon material, a metallic oxide, or polymeric materials is mentioned. As a carbon material, what was prepared in predetermined

temperature and predetermined ambient atmospheres, such as carbon black, such as corks, such as pyrolytic carbon, petroleum coke, or pitch coke, artificial graphites, natural graphites, and acetylene black, glassy carbon, an organic polymeric-materials baking object, or a carbon fiber, is mentioned, for example. In addition, an organic polymeric-materials baking object calcinates organic polymeric materials at the suitable temperature of 500 degrees C or more in an inert gas ambient atmosphere or a vacuum. Moreover, as a metallic oxide, ferrous oxide, ruthenium oxide, or molybdenum oxide is mentioned, and polyacetylene or polypyrrole is mentioned as polymeric materials.

[0075] Considering the material into which lithium is intercalated or deintercalated, the simple substance of the metallic element which can form a lithium and an alloy, or metalloid element, an alloy, or a compound is also mentioned. In addition, the thing which becomes an alloy from two or more sorts of metallic elements in this specification, in addition, what consists of one or more sorts of metallic elements and one or more sorts of metalloid element is included. The solid solution, an eutectic (eutectic mixture), intermetallic compounds, or those things with which it strikes and two or more sorts coexist are in the organization.

[0076] As such a metallic element or metalloid element For example, tin (Sn), lead (Pb), aluminum, an indium (In), Silicon, zinc (Zn), copper (Cu), cobalt (Co), antimony (Sb), A bismuth (Bi), cadmium (Cd), magnesium, boron (B), a gallium (Ga), germanium (germanium), an arsenic (As), silver (Ag), a hafnium (Hf), a zirconium (Zr), and an yttrium (Y) are mentioned. As these alloys or a compound, they are a chemical formula Mae Mbf Lig or a chemical formula Mah Mci Mdj, for example. What is expressed is mentioned. In these chemical formulas, Ma expresses at least one sort in the metallic element which can form a lithium and an alloy, and metalloid element, Mb expresses at least one sort in metallic elements other than a lithium and Ma, and metalloid element, Mc expresses at least one sort of a nonmetallic element, and Md expresses at least one sort in metallic elements other than Ma, and metalloid element. Moreover, the values of e, f, g, h, i, and j are  $e > 0$ ,  $f \geq 0$ ,  $g \geq 0$ ,  $h > 0$ ,  $i > 0$ , and  $j \geq 0$ , respectively.

[0077] Especially, silicon, tin, these alloys, or a compound has desirable especially desirable simple substance of 4B group's metallic element, or metalloid element, alloy, or compound. As for these, the thing of a crystalline substance may also be amorphous.

[0078] The examples of the alloy and compound are LiAl, AlSb, CuMgSb, SiB<sub>4</sub>, SiB<sub>6</sub>, Mg<sub>2</sub>Si, Mg<sub>2</sub>Sn and Ni<sub>2</sub>Si, TiSi<sub>2</sub>, MoSi<sub>2</sub>, and CoSi<sub>2</sub>, NiSi<sub>2</sub>, CaSi<sub>2</sub>, CrSi<sub>2</sub>, Cu<sub>5</sub>Si, and FeSi<sub>2</sub>, MnSi<sub>2</sub>, NbSi<sub>2</sub>, TaSi<sub>2</sub>, VSi<sub>2</sub>, WSi<sub>2</sub>, ZnSi<sub>2</sub>, SiC, Si<sub>3</sub>N<sub>4</sub>, Si<sub>2</sub>N<sub>2</sub>O, SiO<sub>v</sub> (0 < v <= 2), SnO<sub>w</sub> (0 < w <= 2), SnSiO<sub>3</sub>, LiSiO, or LiSnO.

[0079] As the material into which lithium is intercalated or deintercalated, any these one sort or two sorts or more may be mixed, and a lithium may be used.

[0080] The positive electrode 12 contains positive active material, and contains electric conduction agents, such as carbon black or graphite, and binders, such as polyvinylidene fluoride, further if needed.

[0081] As positive active material, the material into which lithium is intercalated or deintercalated, and any these one sort or two sorts or more are used. The examples are metallic sulfide or oxide which does not contain a lithium such as TiS<sub>2</sub> and MoS<sub>2</sub> or V<sub>2</sub>O<sub>5</sub> etc, or lithium composite oxide containing lithium, or NbSe<sub>2</sub> etc. In order to make an energy density high especially, it is preferably lithium composite oxide such as Li<sub>x</sub>MO<sub>2</sub>. The lithium multiple oxide made into a subject is desirable. In addition, one or more kinds of transition-metals elements of M are desirable, and, specifically, its at least one sort in cobalt, nickel (nickel), and manganese (Mn) is desirable. Moreover, x is usually the

value of  $0.05 \leq x \leq 1.10$ . The examples of lithium composite oxide are  $\text{LiCoO}_2$ ,  $\text{LiNiO}_2$ ,  $\text{Li}_x\text{Ni}_y\text{Co}_{1-y}\text{O}_2$ (the value of x and y changes with charge-and-discharge conditions of a cell, and are usually  $0 < x < 1$  and  $0.7 < y \leq 1$ .) or  $\text{LiMn}_2\text{O}_4$  etc.

[0082] In addition, this lithium multiple oxide carries out grinding mixing according to the presentation of a request of the carbonate of a lithium, a nitrate, an oxide or a hydroxide, and the carbonate of transition metals, a nitrate, an oxide or a hydroxide, and is prepared by calcinating in an oxygen ambient atmosphere at the temperature within the limits of  $600^\circ\text{C}$  to  $1000^\circ\text{C}$ .

[0083] The electrolyte 13 contains lithium salt as an electrolyte salt here. Moreover, the electrolyte 13 serves also as the duty as a separator in this rechargeable battery. That is, a negative electrode 11 and a positive electrode 12 are isolated, and a lithium ion is passed, preventing the short circuit of the current by contact of two poles. In addition, you may make it have the separator which is not illustrated between a positive electrode 12 and an electrolyte 13 or into an electrolyte 13 between a negative electrode 11 and an electrolyte 13 if needed. As a separator, there is what consists of a nonwoven fabric made of synthetic resin, such as polytetrafluoroethylene, polypropylene,

or polyethylene, a thing which consists of a ceramic film, or a thing which consists of a porosity thin film.

[0084] The rechargeable battery which has such a configuration acts as follows.

[0085] In this rechargeable battery, if it charges, a lithium ion will be desorbed from a positive electrode 12, and occlusion will be carried out to a negative electrode 11 through an electrolyte 13, for example. If it discharges, a lithium ion will be desorbed from a negative electrode 11, and return occlusion will be carried out to a positive electrode 12 through an electrolyte 13, for example. Here, since the electrolyte 13 contains the 2nd compound, high ionic conductivity is obtained. Moreover, since the 1st high molecular compound is included, high membrane formation nature is obtained, and since at least one sort in the 3rd compound and the 2nd high molecular compound is included, high elasticity is acquired. Therefore, the outstanding cell engine performance is shown.

[0086] Thus, according to the electrolyte 13 concerning the gestalt of this operation, since it was made for at least one sort in the 1st high molecular compound, the 2nd compound, and the 3rd compound and the 2nd high molecular compound to be included, high membrane formation nature, high

ionic conductivity, and high elasticity can be acquired. Therefore, if this electrolyte is used, the electrochemistry device which has the high engine performance can be obtained easily.

[0087] Moreover, since it was made for the 1st compound, the 2nd compound, and the 3rd compound to be included, the 1st compound can be made to construct a bridge easily with an exposure or heating of ultraviolet rays according to the constituent for electrolytes concerning the gestalt of this operation. Therefore, the electrolyte concerning the gestalt of this operation can be obtained easily.

[0088] Furthermore, since it was made to make the 1st compound construct a bridge about the mixture containing the 1st compound, the 2nd compound, and the 3rd compound according to the manufacture approach of the electrolyte concerning the gestalt of this operation, the electrolyte concerning the gestalt of this operation can be obtained easily.

[0089] In addition, since the electrolyte concerning the gestalt of this operation was used according to the rechargeable battery concerning the gestalt of this operation, while being able to manufacture easily, the high cell engine performance and high flexible nature can be obtained.

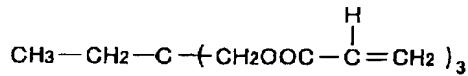
[0090]

[Example] Furthermore, the concrete example of this invention is explained to a detail.

[0091] ), (Example 1-1 to 1-3) Trimethylolpropane triacrylate (TMPTA) having the molecular weight of about 296 shown in formula 28 as the 1st compound, a siloxane derivative having the molecular weight of 29 shown in formula 29 as the 2nd compound, polyethylene oxide (PEO) having the weight average molecular weight of about 1million and lithium trifluoromethane sulfone imide ( $\text{LiN}(\text{CF}_3\text{SO}_2)_2$ ) as an electrolyte salt were prepared.

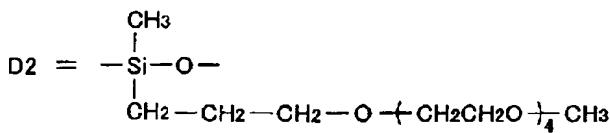
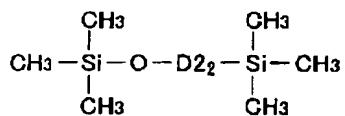
[0092]

[Formula 28]



[0093]

[Formula 29]



[0094] Subsequently, these 1st compounds, the 2nd compound, the 3rd compound, and an electrolyte salt were dissolved in the 2g acetonitrile ( $\text{CH}_3\text{CN}$ ) which performed dehydration processing as a solvent, and it mixed until it became a uniform solution. In that case, in the example 1-1 to 1-3, the addition of the 1st compound, the 2nd compound, and the 3rd compound was changed, respectively, as shown in Table 1. Moreover, it was made, as for the addition of an electrolyte salt, for the mole ratio (lithium atom (Li) / oxygen atom (O)) of a lithium atom and the oxygen atom in an ether unit to be set to 0.06 about all of an example 1-1 to 1-3. Then, add 0.02g 2 and 2-dimethoxy -2 and a 2'-phenyl acetophenone to this mixed solution as a photosensitizer (polymerization initiator), it was made to dissolve in it, and the solution of the constituent for electrolytes was obtained.

[0095]

[Table 1]

|         | 添加量(g)         |              |              | 成膜性         | イオン伝導度<br>(S/cm)     | 折損試験 |
|---------|----------------|--------------|--------------|-------------|----------------------|------|
|         | 第1化合物<br>TMPTA | 第2化合物<br>化29 | 第3化合物<br>PEO |             |                      |      |
| 実施例 1-1 | 1.0            | 10.0         | 10.0         | 良           | $3.1 \times 10^{-5}$ | 折損無  |
| 実施例 1-2 | 2.0            | 10.0         | 10.0         | 良           | $2.4 \times 10^{-5}$ | 折損無  |
| 実施例 1-3 | 2.0            | 10.0         | 5.0          | 良           | $4.6 \times 10^{-5}$ | 折損無  |
| 比較例 1-1 | 0              | 0            | 10.0         | 自己支持性<br>なし | $4.7 \times 10^{-7}$ | 塑性変形 |
| 比較例 1-2 | 0              | 10.0         | 0            | 成膜せず        | $2.5 \times 10^{-4}$ | 試験不可 |
| 比較例 1-3 | 2.0            | 10.0         | 0            | 良           | $1.0 \times 10^{-4}$ | 折損有  |
| 比較例 1-4 | 0              | 10.0         | 10.0         | 成膜せず        | $3.2 \times 10^{-5}$ | 試験不可 |

[0096] The solution of this constituent for electrolytes was applied to homogeneity on the substrate made of a fluororesin after it, in 80°C dry air, it dried for 10 minutes and the acetonitrile was removed. Subsequently, they are 70 mW/cm<sup>2</sup> to a spreading object. Ultraviolet rays were irradiated for 5 minutes and the polymerization reaction was performed about the 1st compound. Then, under the 80-degree C vacuum, it dried for 1 hour, the acetonitrile was removed completely, and the electrolyte of the shape of film with a thickness of 50 micrometers was obtained.

[0097] While investigating membrane formation nature about the obtained electrolyte, it is this electrolyte an area of 1.0cm<sup>2</sup> It clipped to discoid, it put

between the plate electrode made from the stainless steel of a pair, and asked for the ionic conductivity in 25°C by the complex impedance method. Those results are shown in Table 1.

[0098] Moreover, it investigated whether after producing similarly the electrolyte of magnitude with a 2cm x 4cm x thickness of 200 micrometers, bending focusing on a 2cm part from width about an example 1-1 to 1-3, carrying out vacuum enclosure and leaving it with a laminate film, a laminate film is opened and an electrolyte has breakage, or there would be nothing. The result of this breakage trial is also shown in Table 1.

[0099] Except for having not added the 1st compound (TMPTA) and the 2nd compound (siloxane derivative shown in formula 29), but having added only the 3rd compound (PEO) as an example 1-1 of a comparison over this example, the electrolyte was produced like an example 1-1 and 1-2. Moreover, except for having not added the 1st compound (TMPTA) and the 3rd compound (PEO), but having added only the 2nd compound (siloxane derivative shown in formula 29) as an example 1-2 of a comparison over this example, the electrolyte was produced like the example 1-1 to 1-3. Furthermore, except for having not added the 3rd compound (PEO) but having added only the 1st compound

(TMPTA) and the 2nd compound (siloxane derivative shown in formula 29) as an example 1-3 of a comparison over this example, the electrolyte was produced like an example 1-2 and 1-3. In addition, except for having not added the 1st compound (TMPTA) but having added only the 2nd compound (siloxane derivative shown in formula 29), and the 3rd compound (PEO) as an example 1-4 of a comparison over this example, the electrolyte was produced like an example 1-1 and 1-2.

[0100] About the example 1-1 to 1-4 of a comparison as well as an example 1-1 to 1-3, membrane formation nature, ionic conductivity, and the existence of breakage were investigated. The result is shown according to Table 1.

[0101] As shown in Table 1, the film which has free-standing in the example 1-1 of a comparison which does not contain the 1st high molecular compound, 1-2, and 1-4 could not be obtained, and membrane formation was not completed in the example 1-1 of a comparison, and 1-4, either. Moreover, in the example 1-2 of a comparison which does not contain the 2nd compound, ionic conductivity was very low. Furthermore, breakage was seen in the example 1-3 of a comparison which does not contain the 3rd compound. On the other hand, in the example 1-1 to 1-3, membrane formation nature was good, ionic

conductivity was as high as  $2 \times 10$  to 5 or more S/cm, and breakage was not seen, either.

[0102] That is, when making it the 1st high molecular compound (TMPTA), the 2nd compound (siloxane derivative shown in formula 29), and the 3rd compound (PEO) included, it turned out that high membrane formation nature, high ionic conductivity, and high elasticity can be acquired.

[0103] (Example 2-1 to 2-3) As the 3rd compound, it replaced with polyethylene oxide and the weight average molecular weight which has oligo ethylene oxide produced the electrolyte like the example 1-1 to 1-3 except for having used the poly FOSUFAZEN derivative of 700,000 to the side chain shown in formula 26 (J is the repeat number of unit and K= 7). About the example 2-1 to 2-3 as well as an example 1-1 to 1-3, membrane formation nature, ionic conductivity, and the existence of breakage were investigated. The result is shown in Table 2.

[0104]

[Table 2]

|         | 添加量(g)         |              |                           | 成膜性         | イオン伝導度<br>(S/cm)     | 折損試験 |
|---------|----------------|--------------|---------------------------|-------------|----------------------|------|
|         | 第1化合物<br>TMPTA | 第2化合物<br>化29 | 第3化合物<br>ホリファスファゼン<br>誘導体 |             |                      |      |
| 実施例 2-1 | 1.0            | 10.0         | 10.0                      | 良           | $1.2 \times 10^{-4}$ | 折損無  |
| 実施例 2-2 | 2.0            | 10.0         | 10.0                      | 良           | $1.0 \times 10^{-4}$ | 折損無  |
| 実施例 2-3 | 2.0            | 10.0         | 5.0                       | 良           | $1.1 \times 10^{-4}$ | 折損無  |
| 比較例 2-1 | 0              | 0            | 10.0                      | 自己支持性<br>なし | $9.5 \times 10^{-5}$ | 塑性変形 |
| 比較例 1-2 | 0              | 10.0         | 0                         | 成膜せず        | $2.5 \times 10^{-4}$ | 試験不可 |
| 比較例 1-3 | 2.0            | 10.0         | 0                         | 良           | $1.0 \times 10^{-4}$ | 折損有  |

[0105] Except for having not added the 1st compound (TMPTA) and the 2nd compound (siloxane derivative shown in formula 29), but having added only the 3rd compound (poly FOSUFAZEN derivative) as an example 2-1 of a comparison over this example, the electrolyte was produced like an example 2-1 and 2-2. About the example 2-1 of a comparison as well as an example 1-1 to 1-3, membrane formation nature, ionic conductivity, and the existence of breakage were investigated. The result is shown in Table 2 with the example 1-2 of a comparison, and the result of 1-3.

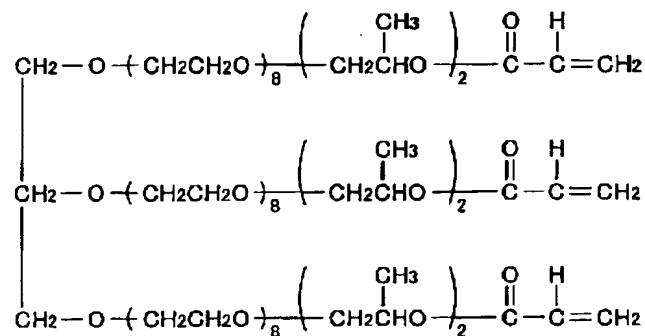
[0106] As shown in Table 2, according to the example 2-1 to 2-3, membrane formation nature was good, ionic conductivity was as high as  $1 \times 10^{-4}$  or more S/cm, and breakage was not seen, either. On the other hand, the film with

free-standing could not be obtained in the example 2-1 of a comparison of only the 3rd compound, but ionic conductivity was also low. That is, even if it used other matter as the 3rd compound, it turned out that high membrane formation nature, high ionic conductivity, and high elasticity can be acquired.

[0107] (Example 3-1 to 3-6) As the 1st compound, it replaced with trimethylolpropane triacrylate and the weight average molecular weight shown in formula 30 produced the electrolyte except for having used the polyethylene oxide / polypropylene oxide thoria KURIRETO of about 8000 (EO-PO-TA) like the example 1-1 to 1-3, and the example 2-1 to 2-3. About the example 3-1 to 3-6 as well as an example 1-1 to 1-3, membrane formation nature, ionic conductivity, and the existence of breakage were investigated. The result is shown in Table 3.

[0108]

[Formula 30]



[0109]

[Table 3]

|         | 添加量 (g)           |              |       |                      | 成膜性         | イオン伝導度 (S/cm)        | 折損試験     |  |  |  |
|---------|-------------------|--------------|-------|----------------------|-------------|----------------------|----------|--|--|--|
|         | 第1化合物<br>EO-PO-TA | 第2化合物<br>化29 | 第3化合物 |                      |             |                      |          |  |  |  |
|         |                   |              | PEO   | ポリフオス<br>ファゼン<br>誘導体 |             |                      |          |  |  |  |
| 実施例 3-1 | 1.0               | 10.0         | 10.0  | 0                    | 良           | $9.0 \times 10^{-5}$ | 折損無      |  |  |  |
| 実施例 3-2 | 2.0               | 10.0         | 10.0  | 0                    | 良           | $1.2 \times 10^{-4}$ | 折損無      |  |  |  |
| 実施例 3-3 | 2.0               | 10.0         | 5.0   | 0                    | 良           | $1.4 \times 10^{-4}$ | 折損無      |  |  |  |
| 実施例 3-4 | 1.0               | 10.0         | 0     | 10.0                 | 良           | $1.7 \times 10^{-4}$ | 折損無      |  |  |  |
| 実施例 3-5 | 2.0               | 10.0         | 0     | 10.0                 | 良           | $1.6 \times 10^{-4}$ | 折損無      |  |  |  |
| 実施例 3-6 | 2.0               | 10.0         | 0     | 5.0                  | 良           | $1.9 \times 10^{-4}$ | 折損無      |  |  |  |
| 比較例 1-1 | 0                 | 0            | 10.0  | 0                    | 自己支持性<br>なし | $4.7 \times 10^{-7}$ | 塑性<br>変形 |  |  |  |
| 比較例 1-2 | 0                 | 10.0         | 0     | 0                    | 成膜せず        | $2.5 \times 10^{-4}$ | 試験<br>不可 |  |  |  |
| 比較例 2-1 | 0                 | 0            | 0     | 10.0                 | 自己支持性<br>なし | $9.5 \times 10^{-5}$ | 塑性<br>変形 |  |  |  |
| 比較例 1-4 | 0                 | 10.0         | 10.0  | 0                    | 成膜せず        | $3.2 \times 10^{-5}$ | 試験不<br>可 |  |  |  |
| 比較例 3-1 | 2.0               | 10.0         | 0     | 0                    | 良           | $2.3 \times 10^{-4}$ | 折損有      |  |  |  |

[0110] Except for having not added the 3rd compound (PEO or the poly FOSUFAZEN derivative), but having added only the 1st compound (EO-PO-TA) and the 2nd compound (siloxane derivative shown in formula 29) as an example 3-1 of a comparison over this example, the electrolyte was produced like an

example 3-2, 3-3, 3-5, and 3-6. About the example 3-1 of a comparison as well as an example 1-1 to 1-3, membrane formation nature, ionic conductivity, and the existence of breakage were investigated. The result is shown in Table 3 with the result of the example 1-1 of a comparison, 1-2, 1-4, and 2-1.

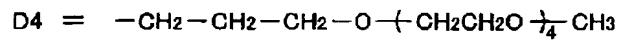
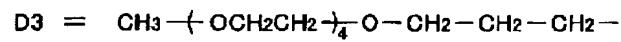
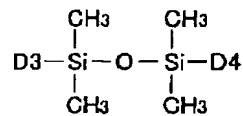
[0111] As shown in Table 3, according to the example 3-1 to 3-3, membrane formation nature was good, ionic conductivity was as high as  $9 \times 10^{-5}$  or more S/cm, and breakage was not seen, either. According to an example 3-4 to 3-6, membrane formation nature was good, ionic conductivity was as high as  $1 \times 10^{-4}$  S/cm, and breakage was not seen, either. On the other hand, breakage was seen in the example 3-1 of a comparison which does not contain the 3rd compound. That is, whether it uses other matter as the 1st compound or used other matter as the 1st high molecular compound, it turned out that high membrane formation nature, high ionic conductivity, and high elasticity can be acquired.

[0112] (An example 4-1 to 4-6, 5-1 to 5-6) It replaced with the siloxane derivative shown in formula 29 as the 2nd compound, and the weight average molecular weight shown in formula 31 produced the electrolyte except for having used the siloxane derivative of 631 like the example 1-1 to 1-3, the

example 2-1 to 2-3, and the example 3-1 to 3-6. Membrane formation nature, ionic conductivity, and the existence of breakage were investigated about an example 4-1 to 4-6, and 5-1 to 5-6 as well as an example 1-1 to 1-3. The result is shown in Table 4 and 5.

[0113]

[Formula 31]



[0114]

[Table 4]

|         | 添加量 (g)        |              |       |                        | 成膜性         | イオン<br>伝導度<br>(S/cm) | 折損<br>試験 |  |  |  |
|---------|----------------|--------------|-------|------------------------|-------------|----------------------|----------|--|--|--|
|         | 第1化合物<br>TMPTA | 第2化合物<br>化31 | 第3化合物 |                        |             |                      |          |  |  |  |
|         |                |              | PEO   | ポリ(オクタ<br>エチレン<br>誘導体) |             |                      |          |  |  |  |
| 実施例 4-1 | 1.0            | 10.0         | 10.0  | 0                      | 良           | $3.2 \times 10^{-5}$ | 折損無      |  |  |  |
| 実施例 4-2 | 2.0            | 10.0         | 10.0  | 0                      | 良           | $2.5 \times 10^{-5}$ | 折損無      |  |  |  |
| 実施例 4-3 | 2.0            | 10.0         | 5.0   | 0                      | 良           | $4.6 \times 10^{-5}$ | 折損無      |  |  |  |
| 実施例 4-4 | 1.0            | 10.0         | 0     | 10.0                   | 良           | $1.2 \times 10^{-4}$ | 折損無      |  |  |  |
| 実施例 4-5 | 2.0            | 10.0         | 0     | 10.0                   | 良           | $1.0 \times 10^{-4}$ | 折損無      |  |  |  |
| 実施例 4-6 | 2.0            | 10.0         | 0     | 5.0                    | 良           | $1.2 \times 10^{-4}$ | 折損無      |  |  |  |
| 比較例 1-1 | 0              | 0            | 10.0  | 0                      | 自己支持性<br>なし | $4.7 \times 10^{-7}$ | 塑性<br>変形 |  |  |  |
| 比較例 2-1 | 0              | 0            | 0     | 10.0                   | 自己支持性<br>なし | $9.5 \times 10^{-5}$ | 塑性<br>変形 |  |  |  |
| 比較例 4-1 | 0              | 10.0         | 0     | 0                      | 成膜せず        | $2.6 \times 10^{-4}$ | 試験<br>不可 |  |  |  |
| 比較例 4-2 | 2.0            | 10.0         | 0     | 0                      | 良           | $1.0 \times 10^{-4}$ | 折損有      |  |  |  |
| 比較例 4-3 | 0              | 10.0         | 10.0  | 0                      | 成膜せず        | $3.3 \times 10^{-5}$ | 試験<br>不可 |  |  |  |

[0115]

[Table 5]

|         | 添加量(g)            |              |       |                      | 成膜性         | イオン伝導度(S/cm)         | 折損試験     |  |  |  |
|---------|-------------------|--------------|-------|----------------------|-------------|----------------------|----------|--|--|--|
|         | 第1化合物<br>EO-PO-TA | 第2化合物<br>化31 | 第3化合物 |                      |             |                      |          |  |  |  |
|         |                   |              | PEO   | ホリファス<br>ファゼン<br>誘導体 |             |                      |          |  |  |  |
| 実施例 5-1 | 1.0               | 10.0         | 10.0  | 0                    | 良           | $9.0 \times 10^{-5}$ | 折損無      |  |  |  |
| 実施例 5-2 | 2.0               | 10.0         | 10.0  | 0                    | 良           | $1.3 \times 10^{-4}$ | 折損無      |  |  |  |
| 実施例 5-3 | 2.0               | 10.0         | 5.0   | 0                    | 良           | $1.4 \times 10^{-4}$ | 折損無      |  |  |  |
| 実施例 5-4 | 1.0               | 10.0         | 0     | 10.0                 | 良           | $1.7 \times 10^{-4}$ | 折損無      |  |  |  |
| 実施例 5-5 | 2.0               | 10.0         | 0     | 10.0                 | 良           | $1.6 \times 10^{-4}$ | 折損無      |  |  |  |
| 実施例 5-6 | 2.0               | 10.0         | 0     | 5.0                  | 良           | $1.9 \times 10^{-4}$ | 折損無      |  |  |  |
| 比較例 1-1 | 0                 | 0            | 10.0  | 0                    | 自己支持性<br>なし | $4.7 \times 10^{-7}$ | 塑性<br>変形 |  |  |  |
| 比較例 2-1 | 0                 | 0            | 0     | 10.0                 | 自己支持性<br>なし | $9.5 \times 10^{-5}$ | 塑性<br>変形 |  |  |  |
| 比較例 4-1 | 0                 | 10.0         | 0     | 0                    | 成膜せず        | $2.6 \times 10^{-4}$ | 試験<br>不可 |  |  |  |
| 比較例 4-3 | 0                 | 10.0         | 10.0  | 0                    | 成膜せず        | $3.3 \times 10^{-5}$ | 試験<br>不可 |  |  |  |
| 比較例 5-1 | 2.0               | 10.0         | 0     | 0                    | 良           | $2.7 \times 10^{-4}$ | 折損有      |  |  |  |

[0116] Except for having not added the 1st compound (TMPTA or EO-PO-TA) and the 3rd compound (PEO or the poly FOSUFAZEN derivative), but having added only the 2nd compound (siloxane derivative shown in formula 31) as an example 4-1 of a comparison over this example, the electrolyte was produced like an example 4-1 to 4-6, and 5-1 to 5-6. Moreover, except for having not added the 3rd compound (PEO or the poly FOSUFAZEN derivative), but having added only the 1st compound (TMPTA) and the 2nd compound (siloxane

derivative shown in formula 31) as an example 4-2 of a comparison over this example, the electrolyte was produced like an example 4-2, 4-3, 4-5, and 4-6. Furthermore, except for having not added the 1st compound (TMPTA or EO-PO-TA), but having added only the 2nd compound (siloxane derivative shown in formula 31), and the 3rd compound (PEO) as an example 4-3 of a comparison over this example, the electrolyte was produced like an example 4-1, 4-2, 5-1, and 5-2. In addition, except for having not added the 3rd compound (PEO or the poly FOSUFAZEN derivative), but having added only the 1st compound (EO-PO-TA) and the 2nd compound (siloxane derivative shown in formula 31) as an example 5-1 of a comparison over this example, the electrolyte was produced like an example 5-2, 5-3, 5-5, and 5-6.

[0117] Membrane formation nature, ionic conductivity, and the existence of breakage were investigated about the example 4-1 to 4-3 of a comparison, and 5-1 as well as an example 1-1 to 1-3. The result is shown in Table 4 or 5 with the example 1-1 of a comparison, and the result of 2-1.

[0118] As shown in Table 4, according to the example 4-1 to 4-3, membrane formation nature was good, ionic conductivity was as high as  $2 \times 10$  to 5 or more S/cm, and breakage was not seen, either. According to an example 4-4 to 4-6,

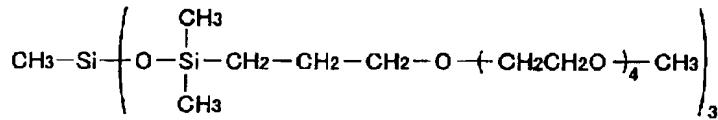
membrane formation nature was good, ionic conductivity was as high as [ more than ] 1x10 to 4 S/cm, and breakage was not seen, either. Moreover, as shown in Table 5, according to the example 5-1 to 5-3, membrane formation nature was good, ionic conductivity was as high as 9x10 to 5 or more S/cm, and breakage was not seen, either. According to an example 5-4 to 5-6, membrane formation nature was good, ionic conductivity was as high as [ more than ] 1x10 to 4 S/cm, and breakage was not seen, either. On the other hand, membranes could not be formed in the example 4-3 of a comparison which does not add the example 4-1 of a comparison of only the 2nd compound, and the 1st compound, but breakage was seen in the example 4-2 of a comparison which does not contain the 3rd compound, and 5-1. That is, even if it used other siloxane derivatives as the 2nd compound, it turned out that high membrane formation nature, high ionic conductivity, and high elasticity can be acquired.

[0119] (An example 6-1 to 6-6, 7-1 to 7-6) It replaced with the siloxane derivative shown in formula 29 as the 2nd compound, and the weight average molecular weight shown in formula 32 produced the electrolyte except for having used the siloxane derivative of 1014 like the example 1-1 to 1-3, the

example 2-1 to 2-3, and the example 3-1 to 3-6. Membrane formation nature, ionic conductivity, and the existence of breakage were investigated about an example 6-1 to 6-6, and 7-1 to 7-6 as well as an example 1-1 to 1-3. The result is shown in Table 6 and 7.

[0120]

[Formula 32]



[0121]

[Table 6]

|         | 添加量 (g)        |              |       |                      | 成膜性         | イオン伝導度<br>(S/cm)     | 折損試験     |  |  |  |
|---------|----------------|--------------|-------|----------------------|-------------|----------------------|----------|--|--|--|
|         | 第1化合物<br>TMPTA | 第2化合物<br>化32 | 第3化合物 |                      |             |                      |          |  |  |  |
|         |                |              | PEO   | ポリフオス<br>ファゼン<br>誘導体 |             |                      |          |  |  |  |
| 実施例 6-1 | 1.0            | 10.0         | 10.0  | 0                    | 良           | $3.2 \times 10^{-5}$ | 折損無      |  |  |  |
| 実施例 6-2 | 2.0            | 10.0         | 10.0  | 0                    | 良           | $2.5 \times 10^{-5}$ | 折損無      |  |  |  |
| 実施例 6-3 | 2.0            | 10.0         | 5.0   | 0                    | 良           | $4.6 \times 10^{-5}$ | 折損無      |  |  |  |
| 実施例 6-4 | 1.0            | 10.0         | 0     | 10.0                 | 良           | $1.2 \times 10^{-4}$ | 折損無      |  |  |  |
| 実施例 6-5 | 2.0            | 10.0         | 0     | 10.0                 | 良           | $1.0 \times 10^{-4}$ | 折損無      |  |  |  |
| 実施例 6-6 | 2.0            | 10.0         | 0     | 5.0                  | 良           | $1.2 \times 10^{-4}$ | 折損無      |  |  |  |
| 比較例 1-1 | 0              | 0            | 10.0  | 0                    | 自己支持性<br>なし | $4.7 \times 10^{-7}$ | 塑性<br>変形 |  |  |  |
| 比較例 2-1 | 0              | 0            | 0     | 10.0                 | 自己支持性<br>なし | $9.5 \times 10^{-5}$ | 塑性<br>変形 |  |  |  |
| 比較例 6-1 | 0              | 10.0         | 0     | 0                    | 成膜せず        | $2.6 \times 10^{-4}$ | 試験<br>不可 |  |  |  |
| 比較例 6-2 | 2.0            | 10.0         | 0     | 0                    | 良           | $1.0 \times 10^{-4}$ | 折損有      |  |  |  |
| 比較例 6-3 | 0              | 10.0         | 10.0  | 0                    | 成膜せず        | $3.3 \times 10^{-5}$ | 試験<br>不可 |  |  |  |

[0122]

[Table 7]

|         | 添加量(g)            |              |       |                      | 成膜性     | イオン伝導度(S/cm)         | 折損試験 |  |  |  |
|---------|-------------------|--------------|-------|----------------------|---------|----------------------|------|--|--|--|
|         | 第1化合物<br>EO-PO-TA | 第2化合物<br>化32 | 第3化合物 |                      |         |                      |      |  |  |  |
|         |                   |              | PEO   | ポリフオス<br>ファゼン<br>誘導体 |         |                      |      |  |  |  |
| 実施例 7-1 | 1.0               | 10.0         | 10.0  | 0                    | 良       | $9.0 \times 10^{-5}$ | 折損無  |  |  |  |
| 実施例 7-2 | 2.0               | 10.0         | 10.0  | 0                    | 良       | $1.3 \times 10^{-4}$ | 折損無  |  |  |  |
| 実施例 7-3 | 2.0               | 10.0         | 5.0   | 0                    | 良       | $1.4 \times 10^{-4}$ | 折損無  |  |  |  |
| 実施例 7-4 | 1.0               | 10.0         | 0     | 10.0                 | 良       | $1.7 \times 10^{-4}$ | 折損無  |  |  |  |
| 実施例 7-5 | 2.0               | 10.0         | 0     | 10.0                 | 良       | $1.6 \times 10^{-4}$ | 折損無  |  |  |  |
| 実施例 7-6 | 2.0               | 10.0         | 0     | 5.0                  | 良       | $1.9 \times 10^{-4}$ | 折損無  |  |  |  |
| 比較例 1-1 | 0                 | 0            | 10.0  | 0                    | 自己支持性なし | $4.7 \times 10^{-7}$ | 塑性変形 |  |  |  |
| 比較例 2-1 | 0                 | 0            | 0     | 10.0                 | 自己支持性なし | $9.5 \times 10^{-5}$ | 塑性変形 |  |  |  |
| 比較例 6-1 | 0                 | 10.0         | 0     | 0                    | 成膜せず    | $2.6 \times 10^{-4}$ | 試験不可 |  |  |  |
| 比較例 6-3 | 0                 | 10.0         | 10.0  | 0                    | 成膜せず    | $3.3 \times 10^{-5}$ | 試験不可 |  |  |  |
| 比較例 7-1 | 2.0               | 10.0         | 0     | 0                    | 良       | $2.7 \times 10^{-4}$ | 折損有  |  |  |  |

[0123] Except for having not added the 1st compound (TMPTA or EO-PO-TA) and the 3rd compound (PEO or the poly FOSUFAZEN derivative), but having added only the 2nd compound (siloxane derivative shown in formula 32) as an example 6-1 of a comparison over this example, the electrolyte was produced like an example 6-1 to 6-6, and 7-1 to 7-6. Moreover, except for having not added the 3rd compound (PEO or the poly FOSUFAZEN derivative), but having added only the 1st compound (TMPTA) and the 2nd compound (siloxane

derivative shown in formula 32) as an example 6-2 of a comparison over this example, the electrolyte was produced like an example 6-2, 6-3, 6-5, and 6-6. Furthermore, except for having not added the 1st compound (TMPTA or EO-PO-TA), but having added only the 2nd compound (siloxane derivative shown in formula 32), and the 3rd compound (PEO) as an example 6-3 of a comparison over this example, the electrolyte was produced like an example 6-1, 6-2, 7-1, and 7-2. In addition, except for having not added the 3rd compound (PEO or the poly FOSUFAZEN derivative), but having added only the 1st compound (EO-PO-TA) and the 2nd compound (siloxane derivative shown in formula 32) as an example 7-1 of a comparison over this example, the electrolyte was produced like an example 7-2, 7-3, 7-5, and 7-6.

[0124] Membrane formation nature, ionic conductivity, and the existence of breakage were investigated about the example 6-1 to 6-3 of a comparison, and 7-1 as well as an example 1-1 to 1-3. The result is shown in Table 6 or 7 with the example 1-1 of a comparison, and the result of 2-1.

[0125] As shown in Table 6, according to the example 6-1 to 6-3, membrane formation nature was good, ionic conductivity was as high as  $2 \times 10$  to 5 or more S/cm, and breakage was not seen, either. According to an example 6-4 to 6-6,

membrane formation nature was good, ionic conductivity was as high as  $1 \times 10^{-4}$  S/cm, and breakage was not seen, either. Moreover, as shown in Table 7, according to the example 7-1 to 7-3, membrane formation nature was good, ionic conductivity was as high as  $9 \times 10^{-5}$  or more S/cm, and breakage was not seen, either. According to an example 7-4 to 7-6, membrane formation nature was good, ionic conductivity was as high as  $1 \times 10^{-4}$  S/cm, and breakage was not seen, either. On the other hand, membranes could not be formed in the example 6-3 of a comparison which does not add the example 6-1 of a comparison of only the 2nd compound, and the 1st compound, but breakage was seen in the example 6-2 of a comparison which does not contain the 3rd compound, and 7-1. That is, even if it used other siloxane derivatives as the 2nd compound, it turned out that high membrane formation nature, high ionic conductivity, and high elasticity can be acquired.

[0126] In addition, the same result can be obtained even if it uses other matter which fills with the above-mentioned example the conditions explained in the gestalt of the above-mentioned implementation although an example was given and the 1st compound, the 2nd compound, the 3rd compound, and an electrolyte salt were explained concretely.

[0127] As mentioned above, although the gestalt and example of operation were given and this invention was explained, this invention is not limited to the gestalt and example of the above-mentioned implementation, and is variously deformable. For example, as long as it fulfills the conditions explained in the gestalt of the above-mentioned implementation, you may make it use other matter in the gestalt of the above-mentioned implementation, although an example was given and the 1st compound, the 2nd compound, the 3rd compound, and an electrolyte salt were explained concretely.

[0128] Moreover, in the gestalt of the above-mentioned implementation, although the rechargeable battery using a lithium was explained and the case where lithium salt was used as an electrolyte salt was explained in the above-mentioned example, this invention is applicable about the electrolyte and rechargeable battery which used other electrolyte salts, such as sodium salt or a calcium salt, similarly.

[0129] Furthermore, in the gestalt of the above-mentioned implementation, although the rechargeable battery of a paper mold was explained, this invention is applicable about the thing of other configurations, such as telescopic [ which has a carbon button mold, a coin mold, a square shape, or spiral structure ],

similarly.

[0130] In addition, in the gestalt of the above-mentioned implementation, although the case where the electrolyte of this invention was used for a rechargeable battery was explained, the constituent for an electrolyte and electrolytes of this invention can be used about other cells, such as a primary cell. Furthermore, it can also use for other electrochemistry devices, such as a capacitor, a capacitor, or an electrochromic element. In addition, for example, when using the electrolyte of this invention for a capacitor etc., basic group salts, such as ammonium salt, can also be used as an electrolyte salt.

[0131]

[Effect of the Invention] Since it was made for the 1st compound, the 2nd compound, and the 3rd compound to be included according to the constituent for electrolytes given in any 1 term of claim 1 thru/or claim 11 as explained above, the electrolyte of this invention can be easily obtained according to bridge formation. That is, the electrolyte which has high membrane formation nature, high ionic conductivity, and high elasticity can be obtained easily.

[0132] Moreover, according to the electrolyte given in any 1 term of claim 12 to claim 22, since it was made for at least one sort in the 1st high molecular

compound, the 2nd compound, and the 3rd compound and the 2nd high molecular compound to be included, high membrane formation nature, high ionic conductivity, and high elasticity can be acquired. Therefore, if this electrolyte is used, the electrochemistry device which has the high engine performance can be obtained easily.

[0133] Furthermore, since it was made to make the 1st compound construct a bridge about the mixture containing the 1st compound, the 2nd compound, and the 3rd compound according to the manufacture approach of an electrolyte according to claim 23 or 24, the electrolyte concerning the gestalt of this operation can be obtained easily.

[0134] In addition, since the electrolyte of this invention was used, while being able to manufacture easily according to the cell given in any 1 term of claim 25 to claim 35, the high cell engine performance and high flexible nature can be obtained.

[Brief Description of the Drawings]

[FIG. 1] It is a sectional view showing the configuration of the rechargeable battery using the electrolyte concerning the gestalt of 1 operation of this

invention.

[Description of Notations]

11 negative electrode; 12 positive electrode; 13 electrolyte; 14 Insulating packing; 15, 16 outer member